Neutron Generator at Hiroshima University for Use in Radiobiology Study

S. ENDO*,†, M. HOSHI†, H. TAUCHI†, S. TAKEOKA†, K. KITAGAWA†,
S. SUGA†, N. MAEDA†, K. KOMATSU†, S. SAWADA†,
E. IWAMOTO‡, S. SAKAMOTO‡, K. TAKEYAMA‡
AND M. OMURA‡

†Research Institute for Radiation Biology and Medicine, Hiroshima University,
Kasumi 1-2-3, Minami-ku, Hiroshima 734, Japan
‡Nisshin High Voltage Co. Ltd., Souja 2121, Maebashi, Gunma 371, Japan

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A neutron generator (HIRRAC) for use in radiobiology study has been constructed at the Research Institute for Radiation Biology and Medicine, Hiroshima University (RIRBM). Monoenergetic neutrons of which energy is less than 1.3 MeV are generated by the $^7$Li(p,n)$^7$Be reaction at proton energies up to 3 MeV. The protons are accelerated by a Schenkel-type accelerator and are bombarded onto the $^7$Li-target. An apparatus for the irradiation of biological material such as mice, cultured cells and so on, was designed and will be manufactured. Neutron and gamma-ray dose rates were measured by paired (TE-TE and C-CO$_2$) ionization chambers. Contamination of the gamma ray was less than about 6% when using 10-μm-thick $^7$Li as a target. Maximum dose rates for the tissue equivalent materials was 40 cGy/min at a distance of 10 cm from the target. Energy distributions of the obtained neutrons have been measured by a $^3$He-gas proportional counter. The monoenergetic neutrons within an energy region from 0.1 to 1.3 MeV produced by thin $^7$Li or $^7$LiF targets had a small energy spread of about 50 keV (1σ width of gaussian). The energy spread of neutrons was about 10% or less at an incident proton energy of 2.3 MeV. We found that HIRRAC produces small energy spread neutrons and at sufficient dose rates for use in radiobiology studies.

1. INTRODUCTION

To study the radiobiological effects of neutrons, we have constructed a neutron irradiating system, the Hiroshima University Radiobiological Research Accelerator (HIRRAC), which is operated at high proton beam currents of 1 mA. The available neutron energy range is from 0.05 to 1.3 MeV by using $^7$Li and $^7$LiF targets. Dose rate, gamma ray contaminations and energy distributions of the neutron field obtained by HIRRAC were measured for use in radiobiology studies.

*Author for correspondence.
Concerning the studies of radiobiological effects for different energy neutrons, we are interested in the neutron doses of atomic bomb (A-bomb) survivors in Hiroshima and Nagasaki, which is estimated by Dosimetric System 1986 (DS86) (RERF 1987). Research groups at RIRBM and the Faculty of Engineering, Hiroshima University are studying inconsistencies between A-bomb-neutron induced activation and calculated yields based on DS86 (Hasai et al. 1987, Hoshi et al. 1989, Shizuma et al. 1989). They are questioning both the neutron dose and energy spectrum in Hiroshima estimated by DS86, since peaks of neutron energy spectra are about 0.5 MeV in Hiroshima and about 2 MeV in Nagasaki from DS86 calculated results (RERF 1987). This difference may correspond to that of relative biological effectiveness (RBE). Radiation effects for survivors such as chromosomal aberration frequencies at a given dose in Hiroshima is larger than that of Nagasaki (Awa 1991). Therefore, neutron-energy dependencies of RBEs may be important in the analyses concerning risk of radiation based on Hiroshima and Nagasaki.

On the other hand, Miller et al. suggested that RBEs for the neoplastic transformation in C3H10T1/2 mouse cell had a maximum value at a neutron energy of about 0.35 MeV (Miller et al. 1989). However, their data have relatively large statistical fluctuations, so we are planning to irradiate biological materials to obtain RBEs.

A high dose rate of a monochromatic neutron field are useful to study the neutron-energy dependency of the biological effects, and also, for other radiobiology studies to consider the basic mechanisms of the effects of neutrons.

### 2. MATERIALS AND METHODS

#### 2.1. HIRRAC

HIRRAC consists of a Schenkel type 3MV ion accelerator (HN-3000BL) manufactured by Nisshin High Voltage Co. Ltd., and target assemblies for neutron generation. Thin $^7$Li or $^7$LiF targets were prepared by evaporation onto 0.5-mm-thick copper disks. The copper disks were mounted on the end of a beam duct and were cooled with water to prevent heating by proton beam currents. Schematic diagrams of HIRRAC are shown in Figure 1(a) and (b). Ion beams are produced in a RF-ion-source generator and is accelerated through a Schenkel-type high-voltage acceleration tube. Nominal beam currents at maximum for protons are 1 mA. Beam parameters of the 3MV ion accelerator are summarized in Table 1.

The energy of the accelerated beam is analyzed by a bending magnet (BM-1), whose magnetic field is monitored by a nuclear magnetic resonance method (NMR). The analyzed beam is focused by a quadrupole magnet (QM-1) and extracted to the experimental room. The beam is distributed to four beam lines in the experimental room by using a switching magnet (SM). For neutron irradiation, there are two beam lines, a horizontal beam line (H-Line) and a vertical one (V-Line) using $^7$Li target. The V-line is suitable to irradiate cells in monolayer in medium. The others are for the use of $^3$H target (T-Line) and quantitative analysis with a proton-induced x-ray emission (PIXE) method. The V-Line is settled in a 2-m-deep cylindrical well as shown in Figure 1(b). The vertical ion beam of the V-Line bombards the $^7$Li target in the
The $^7\text{Li}$, $^7\text{LiF}$, $^9\text{Be}$ (for H-Line and V-Line) and $^3\text{H}$ targets (T-Line) are designed to produce neutrons at various energies (50 keV−15 MeV). The optimizing calculation for the target thickness was performed to produce in a condition of neutron energy spread of less than 10% ($\Delta E/E \leq 0.1$) at an acceleration energy of 2.3 MeV. The energy loss per unit of $^7\text{Li}$ is about 8 keV/\(\mu\text{m}\).

**Figure 1.** Schematic diagrams of HIRRAC. (a) top view and (b) side view.
Therefore, 15-μm-thick ⁷Li corresponds to about 120 keV energy loss. Similarly, the same energy losses as 120 keV are produced by about 5-μm-thick ⁷LiF and about 1-μm-thick ⁹Be. Such targets are made by vacuum evaporation of target materials onto copper disk. The thicknesses of the targets are controlled by the evaporated weight of each target material. In this paper, we present data obtained by a 4-μm-thick ⁷LiF target and about 10-μm-thick ⁷Li targets.

2.2 Proton energy calibration

A proton energy from the ion accelerator is calibrated against four absolute energies; (1) the reaction threshold 1.881 MeV for the ⁷Li(p,n)⁷Be reaction, and (2)—(4) resonances at 0.874, 0.935 and 1.375 MeV, respectively, for the ¹⁹F(p,γ)¹⁶O reaction. The neutrons produced by the ⁷Li(p,n)⁷Be reaction of which threshold energy is 1.881 MeV, were measured by a BF₃ counter and the resonance gammarays, 0.874, 0.935 and 1.375 MeV, were measured by a NaI scintillation counter. Each energy is corresponded to the magnetic field strength of BM-1 (B), which is measured by nuclear magnetic resonance (NMR).

2.3. Neutron spectrometer

Neutron energy was measured by a ³He gas proportional counter coupled with a preamplifier (FAST NEUTRON SPECTROMETER MODEL FNS-1, Jordan Valley Applied Radiation Ltd.). Output signals were magnified by an amplifier module (ORTEC 575A) and were fed to a multi-channel-buffer module (ORTEC 918A). The pulse height is analyzed by a personal computer (NEC PC9801FA/A7). The proportional counter is a gas chamber filled with ³He gas at high pressure (10 atm, the size of the active volume is 5 cm in diameter and 15 cm in length), which are thick enough to stop the low-energy charged particles in the counter. The proton ranges of these energies are about 5 mm or less and the triton ranges are shorter than those of protons. Therefore, output pulse height is proportional to energy deposit of protons and tritons which are produced by the ³He(n,p)t reaction. The best energy resolution (31 keV) was obtained from this system when test pulses from a pulse generator (NAIG E-512) were applied for input signals.

The relative detector-efficiency of the counter is estimated by fission neutrons from the ²⁵²Cf source at RIRBM (Hoshi et al. 1988). As for the energy distribution of the ²⁵²Cf neutron, we assumed the Maxwell Boltzman distribution,

\[ f(E) = E^{\frac{11}{2}} \exp(-E/E_0) \]

The Table 1. Beam parameters of 3MV-ion acceleator.

<table>
<thead>
<tr>
<th></th>
<th>Maximum</th>
<th>Minimum</th>
</tr>
</thead>
<tbody>
<tr>
<td>H⁺-beam current</td>
<td>1 mA</td>
<td>&lt; 10 nA</td>
</tr>
<tr>
<td>D⁺-beam current</td>
<td>700 μA</td>
<td>&lt; 10 nA</td>
</tr>
<tr>
<td>He⁺-beam current</td>
<td>300 μA</td>
<td>&lt; 10 nA</td>
</tr>
<tr>
<td>Acceleration energy</td>
<td>3.0 MV</td>
<td>&lt; 0.2 MV</td>
</tr>
<tr>
<td>Energy Stability</td>
<td>±0.01 MV</td>
<td></td>
</tr>
</tbody>
</table>

Therefore, 15-μm-thick ⁷Li corresponds to about 120 keV energy loss. Similarly, the same energy losses as 120 keV are produced by about 5-μm-thick ⁷LiF and about 1-μm-thick ⁹Be. Such targets are made by vacuum evaporation of target materials onto copper disk. The thicknesses of the targets are controlled by the evaporated weight of each target material. In this paper, we present data obtained by a 4-μm-thick ⁷LiF target and about 10-μm-thick ⁷Li targets.

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The relative detector-efficiency of the counter is estimated by fission neutrons from the ²⁵²Cf source at RIRBM (Hoshi et al. 1988). As for the energy distribution of the ²⁵²Cf neutron, we assumed the Maxwell Boltzman distribution,
where \( E_0 \) is assumed to be 1.42 MeV (Barnard 1965). The relative detector-efficiency by different neutron energies of the detector is obtained as ratios of the measured spectrum to that from equation 1. Figure 2 shows the relative detector-efficiency and a calculated relative efficiency according to a Monte Carlo simulation. Details of the simulation are described in the Appendix. It is shown that the measured detector-efficiency is well reproduced by the calculated efficiency, as shown in Figure 2(a). Moreover, the neutron energy spectra generated by HIRRAC were checked by the Monte Carlo calculation, as shown in Figure 2(b). Therefore, we estimated neutron energy distributions by using the calculated efficiency.

![Figure 2. (a) Relative detector-efficiencies obtained from a measurement and a calculation. Small dots are experimentally obtained relative detector-efficiencies for the \( ^{252}\text{Cf} \)-fission neutron source and the solid curve is the calculation. (b) The generated neutron spectrum at proton energy 2.8 MeV is reproduced well by the calculation.](image)

### 2.4. Ionization chamber

Neutron and gamma-ray kerma rates from HIRRAC are obtained with the paired ionization chambers. The chambers are IC-17 (TE-TE chamber) and IC-17G (C-CO\(_2\) chamber). Calibration procedures and the other procedures of the measurements have already been described elsewhere (Hoshi et al. 1988). The neutron sensitivities of these chambers are assumed to be \( k_T = 0.98 \) and \( k_U = 0.08 \) (Kawashima et al. 1978, ICRU Report 27 1978), respectively. These factors were used to determine the \( ^{252}\text{Cf} \)-neutron kerma. The neutron kerma difference between the assumptions of \( k_U = 0.08 \) and of \( k_U = 0 \) are 5% or less (ICRU 1987, Waterman et al. 1979).

### 3. RESULTS AND DISCUSSION

The threshold energy of 1.881 MeV is determined by a BF\(_3\)-counter, as shown in Figure 3(a). The threshold is clearly seen at a magnetic field strength of 404.9 mT. The resonance gamma-rays, 0.874, 0.935 and 1.375 MeV, are measured by a NaI scintillation counter, as shown
in Figure 3(b) and (c). The resonance spectra have a width of 140 keV in full width at half maximum (FWHM) due to the $^7$LiF target thickness. The magnetic field strengths of 274.84, 284.64 and 346.02 mT which correspond to each resonance energy is obtained by gamma-ray yields in the spectra as shown in Figure 3(b) and (c). The four energy points are plotted in Figure 4 as a function of the square of the magnetic field strength ($B^2$) of BM-1, which is obtained by NMR. The absolute energy $T_p$ is expressed as a function of $B^2$ and a fitting procedure was performed. The result is expressed as $T_p = 0.0059 + 11.474 B^2$ MeV. Moreover, the linearity of the acceleration energy was also calibrated by magnetic field strength of BM-1. The accuracy of the absolute proton energy is within 0.3%.

The result of the measurement are listed in Table 2 for neutron energies of 0.11, 0.37, 0.57

<table>
<thead>
<tr>
<th>Neutron energy (MeV)</th>
<th>Spread (MeV) in $\sigma$</th>
<th>$\Delta E/E$</th>
<th>Dose rate (Gy/min·μA)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.99</td>
<td>0.050</td>
<td>0.05</td>
<td>$3.09 \times 10^{-4}$</td>
</tr>
<tr>
<td>0.57</td>
<td>0.048</td>
<td>0.09</td>
<td>$4.46 \times 10^{-4}$</td>
</tr>
<tr>
<td>0.37</td>
<td>0.035</td>
<td>0.09</td>
<td>$9.01 \times 10^{-5}$</td>
</tr>
<tr>
<td>0.11</td>
<td>—</td>
<td>—</td>
<td>$4.35 \times 10^{-5}$</td>
</tr>
</tbody>
</table>
and 0.99 MeV. The maximum dose rate is about 40 cGy/min at a proton energy of $T_p=2.3$ MeV with a beam current of 1 mA. The measured values of gamma-ray contamination for $^7$Li and $^7$LiF targets for proton energies of 0.37, 0.57 and 0.99 MeV are listed in Table 2. The gamma-ray contaminations for the $^7$LiF target are high (Table 3). These gamma rays mainly are induced by the $^{19}$F(p,α)$^{16}$O reaction, of which gamma-ray energies are 6.14, 6.92 and 7.12 MeV. The $^7$LiF target is easier to handle compared with the $^7$Li target because lithium fluoride is a stable inorganic compound. Lithium fluoride is used when gamma-ray contamination is not effective in radiobiology studies. In most radiobiology studies, experiments are carried out under a condition of low gamma-ray contamination, about 5% or less with a $^7$Li target.

Table 3. Percentage of gamma-ray contaminations for 10 μm $^7$Li, and 4 μm $^7$LiF targets at proton energies of 2.8, 2.6 and 2.4 MeV. For comparison, gamma-ray contaminations of $^{252}$Cf-fission neutrons are also listed. The errors are indicated in parentheses.

<table>
<thead>
<tr>
<th>proton energy (MeV)</th>
<th>2.4</th>
<th>2.6</th>
<th>2.8</th>
</tr>
</thead>
<tbody>
<tr>
<td>10 μm $^7$Li</td>
<td>5.3 (1.2)</td>
<td>1.3 (0.3)</td>
<td>4.6 (1.0)</td>
</tr>
<tr>
<td>4 μm $^7$LiF</td>
<td>44.8 (1.5)</td>
<td>31.3 (5.3)</td>
<td>37.2 (12)</td>
</tr>
<tr>
<td>$^{252}$Cf</td>
<td></td>
<td></td>
<td>34 (1.0)</td>
</tr>
</tbody>
</table>

Figure 4. Calibration of the absolute energy of protons against a magnetic field. The absolute energy obtained in Figure 3 are plotted as a function of the magnetic field. The straight line is obtained by fitting as explained in the text.
The neutron energy spectrum at proton energies of 2.0, 2.2, 2.4 and 2.8 MeV (corresponding to the neutron energies of 0.11, 0.37, 0.57 and 0.99 MeV, respectively) is measured by the gas proportional counter shown in Figure 5. In this case, a thin $^7\text{Li}$ target (10 $\mu$m) was used. The energy spectra are clearly seen at the proton energy from 2.2 to 2.8 MeV. At the proton energy 2.0 MeV, the fast neutron peak, which should be observed at 0.11 MeV, is hidden in the thermal neutron peak. The relationships between neutron energies and energy spreads are also summarized in Table 2. In the energy region from 0.37 to 0.99 MeV, almost no energy dependence was seen and its spread was about 50 keV. Each spectrum is corrected by the detector-efficiency, and the neutron energy fluences for proton energies of 2.0, 2.2, 2.4 and 2.8 MeV (corresponding to the neutron energy of 0.11, 0.37, 0.57 and 0.99 MeV, respectively) are shown in Figure 6.

Figure 5. The raw neutron spectra measured by a $^3\text{He}$-gas proportional counter at incident proton energies of 2.0, 2.2, 2.4 and 2.8 MeV.

Good quality monochromatic neutrons were produced from HIRRAC, as seen in Figure 6. When the HIRRAC is used, studies concerning the energy dependence of RBEs can be performed within 10% resolution by using the 10-$\mu$m-thick $^7\text{Li}$ targets.

The thermal neutron contributions in absorbed doses were estimated from the spectrum of the $^3\text{He}$-gas proportional counter, the cross section of the $^3\text{He}(n,p)t$ reaction and the kerma. Absorbed dose ratios of thermal neutrons to fast neutrons at a distance of 10 cm is estimated to be less than 2% for a neutron-energy region from 0.1 to 1.3 MeV. A precise study for the thermal contribution is in progress.
Figure 6. Relative neutron energy fluences from HIRRAC are shown. The spectra are obtained using the detection efficiencies which are calculated by Monte Carlo simulation.

4. CONCLUSION

We have constructed the HIRRAC irradiation system with small neutron energy spread, $\Delta E/E \leq 10\%$ in the energy region from 0.37 to 1.3 MeV system at a high-dose rate up to 40 cGy/min. According to the present study, we found that the energy dependence on RBEs will be tested under the condition within 10% resolution when the 10-$\mu$m-thin $^7$Li targets were used.

5. ACKNOWLEDGMENT

The authors would like to thank Drs. K. Kobayashi and T. Kobayashi at Research Reactor Institute, Kyoto University for their advice. They are also grateful to Miss K. Kumamoto, Administration Office of the Radiation Facilities, RIRBM, Hiroshima University, for her help.

REFERENCES


Appendix: Monte Carlo calculation

Stopping power for protons in materials have a maximum value at about 0.1 MeV. Such a large energy loss of protons not only shifts the energy of the generated neutrons of the \( p, n \) reaction, but also worsens the energy spread of the generated neutrons. To obtain high-dose rate neutrons, thicker targets are better within an acceptable energy spread for radiobiology studies. To find an optimized condition between dose rates and energy spreads, a Monte Carlo simulation was performed. Five assumptions were used in the calculation; (1) 10 keV stability of the ion-acceleration energy, (2) the energy losses in \(^7\)Li targets is calculated assuming the Bethe-Thomas formula with shell corrections (Northcliffe and Shilling 1970, ICRU 1984), (3) the kinematics for the \(^7\)Li(p,n)\(^7\)Be, (4) scattering of neutron at the target holder and cooling water, (5) detector responses. For (2), probabilities \( P_n \) of neutron generations as functions of the density \( \rho \), the cross section for the \(^7\)Li(p,n)\(^7\)Be reaction \( \sigma \) and a depth of reaction points \( x \) are small enough, so approximation of the equation \( P_n = \rho \sigma x \) is valid. In this code, the probability of proton energy loss is assumed to be constant and probabilities of neutron generations are proportional to the cross section of the \(^7\)Li(p,n)\(^7\)Be reaction. For (4), an average scattering probability in the target assembly materials of 1-mm Cu, 3-mm H\(_2\)O and 3-mm stainless steel for \( (n,n') \) are used here. The scattered neutron energy is calculated with the kinematics assuming uniform angular distributions. For (5), generated neutrons are incident to the detector and interact with \(^3\)He nuclei according to the \(^3\)He(n,p)t or the \(^3\)He(n,n)\(^3\)He reactions below 2 MeV of neutron energies. The probability of each interaction (denoted by \( P(n,p) \) and \( P(n,n) \)) are assumed by

\[
P(n, p) = \frac{\sigma(n, p)}{\sigma(n, p) + \sigma(n, n)} = \left[ 1 - \exp \left\{ - n_{3He} \rho_{3He} (\sigma(n, p) + \sigma(n, n)) \right\} \right]
\]

\[
P(n, n) = \frac{\sigma(n, n)}{\sigma(n, p) + \sigma(n, n)} = \left[ 1 - \exp \left\{ - n_{3He} \rho_{3He} (\sigma(n, p) + \sigma(n, n)) \right\} \right]
\]

where \( n_{3He} \) is a number of \(^3\)He nuclei. Total energy depositions of incident neutron were calculated as follows: (1) In the \(^3\)He(n,p)t reaction which were taken from Neutron Cross Sections Volume 2 Academic Press, Inc. (edited by V. McLane, C. L. Dunford and P. F. Rose): Produced protons and tritons are stopped in a high pressure \(^3\)He-gas (10 atm) and deposite the total energy to the detector. (2) the \(^3\)He(n,n')\(^3\)He reaction followed by \(^3\)He(n',p)t reaction: In this case, the scattered neutron \( (n') \) re-interacts according to the \(^3\)He(n',p)t reaction before passing through the detector. The energies of protons and tritons are calculated again. Total deposite energies are obtained as the sum of energies for the recoil \(^3\)He-nuclei, the emitted protons and tritons. Events for the \(^3\)He(n,p)t reaction have depositions biased with 764 keV of Q-value in addition to the incident neutron energy. (3) the \(^3\)He(n,n')\(^3\)He reaction: This category does not follow the \(^3\)He(n,p)t reaction and does not include the scattered neutrons passing thorough the detector. The energies of scattered neutrons are missing energies. The recoil \(^3\)He
and the scattered neutron energies are calculated by the kinematics. The recoil $^3$He are stopped in the detector, and the energy is deposited by the $^3$He nuclei. Events for the $^3$He(n, n')$^3$He reaction have a broad structure from zero energy to about three-fourths of the neutron energy.

The detector response calculation is tested by using the $^{252}$Cf-fission neutron source and the monochromatic neutron generated at 2.8 MeV. The measured detector-efficiencies are reproduced by the calculation within an accuracy of about 10%.